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Neutronics and material attractiveness for PWR thorium systems using monte carlo techniques

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ABSTRACT

Thorium (Th) oxide fuel offers a significant advantage over traditional low-enriched uranium and mixed uranium/plutonium oxide (MOX) fuel irradiated in a Light Water Reactor. The benefits of using thorium include the following: 1) unlike depleted uranium, thorium does not produce plutonium, 2) thorium is a more stable fuel material chemically than LEU and may withstand higher burnups, 3) the materials attractiveness of plutonium in Th/Pu fuel at high burnups is lower than in MOX at currently achievable burnups, and 4) thorium is three to four times more abundant than uranium. This paper quantifies the irradiation of thorium fuel in existing Light Water Reactors in terms of: 1) the percentage of plutonium destroyed, 2) reactivity safety parameters, and 3) material attractiveness of the final uranium and plutonium products. The Monte Carlo codes MCNP/X and the linkage code *Monteburns* were used for the calculations in this document, which is one of the first applications of full core Monte Carlo burnup calculations. Results of reactivity safety parameters are compared to deterministic solutions that are more traditionally used for full core computations.

Thorium is fertile and leads to production of the fissile isotope ^{233}U, but it must be mixed with enriched uranium or reactor-/weapons-grade plutonium initially to provide power until enough ^{233}U builds in. One proposed fuel type, a thorium-plutonium mixture, is advantageous because it would destroy a significant fraction of existing plutonium while avoiding the creation of new plutonium. ^{233}U has a lower delayed neutron fraction than ^{235}U and acts kinetically similar to ^{239}Pu built in from ^{238}U. However, as with MOX fuel, some design changes may be required for our current LWR fleet to burn more than one-third a core of Th/Pu fuel and satisfy reactivity safety limits. The calculations performed in this research show that thorium/plutonium fuel can destroy up to 70% of the original plutonium per pass at 47 GWd/MTU, whereas only about 30% can be destroyed using MOX. Additionally, the materials attractiveness of the final plutonium product of irradiated plutonium/thorium fuel is significantly reduced if high burnups (~94 GWD/MTU) of the fuel can be attained.

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1. Introduction

The United States (US) primarily uses uranium (U) oxide as fuel in its commercial Light Water Reactors (LWRs). However, other countries irradiate materials such as plutonium (Pu) from spent nuclear fuel and/or thorium as part of their nuclear fuel cycle. The benefits of thorium are given in detail below.

1) Less plutonium builds up in Th/LEU than in LEU fuel and more net plutonium can be destroyed in Th/Pu than in MOX fuel because there is less ^{238}U present initially.
2) The materials attractiveness of plutonium in Th/Pu at high burnups (~94 MWt d/kgHM) is lower than in MOX at currently achievable burnups (~47 MWt d/kgHM).
3) Thorium is three to four times more abundant than uranium, offering economic incentives both from mining and from reduced enrichment needs.
4) Thorium is a more stable fuel material chemically than LEU, so it may withstand higher burnups, especially if a new cladding type is developed.

Cladding less oxidizing than zircaloy such as oxide dispersion-strengthened steels or silicon carbide potentially may be able to handle longer irradiation times than currently attained in PWRs. Higher burnups provide longer reactor lifetimes, superior plutonium consumption rates, and reduced material attractiveness of

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the product. The chemical stability of Th is indeed a nonproliferation advantage, which makes thorium fuel harder to separate if reprocessing is ever desired.

Thorium fuel cycles were examined in the beginning of the nuclear era in the 1950s but were not widely implemented, primarily because of the success of uranium fuel cycles. For countries such as the United States with a large supply of uranium and electricity from non-nuclear sources, a new nuclear fuel cycle has not yet become economical. However, in other countries, such as India, with a large thorium reserve and fewer uranium and other resources, thorium fuel cycles are being more actively pursued. To use thorium fuel in a reactor, it must be mixed initially with a fissile material such as enriched uranium or plutonium until it produces enough ^{233}U to sustain criticality. A mixture of thorium and plutonium fuel would reduce the amount of uranium mining and enrichment necessary in the nuclear fuel cycle. Based upon uranium enrichment capacities worldwide, Table 1 shows examples of how enrichment capability could be reduced using thorium-plutonium fuel instead of uranium (Makhijani et al., 2004). In fact, the use of thorium-plutonium fuel instead of traditional uranium–plutonium mixed oxide fuel would increase the overall destruction versus production rate of fissile material. With the future of a permanent used fuel repository unclear, a thorium fuel cycle could help reduce the plutonium and thus americium and curium necessary to be disposed.

The first set of calculations performed in this research focused on neutronic burnup and kinetic safety of solely a Pressurized Water Reactor (PWR), which served as the most simplified model for comparing burnup parameters for various fuel types. It is recognized that a Boiling Water Reactor (BWR) has the potential of burning thorium-based fuel more thoroughly than a PWR because it has varying void fractions in the core axially that can optimize performance (Bjork et al., 2009a and 2009b). In the past, full core simulations of neutron transport and irradiation in PWRs have primarily been performed using deterministic techniques because a full core model required too much memory to model in detail using the Monte Carlo transport code MCNP. However, advancement of both Monte Carlo techniques and computer hardware have made more detailed calculations possible (Fensin, 2009). The results for the irradiation of both thorium- and uranium-based fuels presented in this paper were obtained using Monte Carlo burnup techniques and represent a new era of computational results. Reactor kinetic safety results are compared to values from deterministic techniques, showing the validity of the calculations.

Finally, the burnup results were fed as input into material attractiveness calculations of the final uranium and plutonium products of the irradiated fuel. Material attractiveness is but one of several factors that determine proliferation risk. Material attractiveness is a small factor in the complex decision process used by policy makers for global nuclear fuel cycle decisions. Other factors include:

- Proliferation resistance is that characteristic of a nuclear energy system (NES) that impedes the diversion or undeclared production of nuclear material or misuse of technology by the *Host State* seeking to acquire nuclear weapons or other nuclear explosive devices.
- Physical protection is that characteristic of an NES that impedes the theft of materials suitable for nuclear explosives or radiation dispersal devices (RDDs) and the sabotage of facilities and transportation by *sub-national entities and other non-Host State adversaries*.
- Material control and accounting plays a role against both *sub-national* threats (through the *Host State* system of accounting and control) and *Host State* threats (through IAEA verification of material accountancy information).

Thus, what is analyzed in this document only represents a small, but nevertheless, important piece of quantifying proliferation risk.

Table 1
Approximate reductions in uranium enrichment requirements by deploying full PWR cores of thorium–plutonium fuel (MTSWU is metric tons of separative work units).

Global Th deployment scenarios	Enriched U savings (tonnes/yr)	Excess enrichment capacity (MTSWU/year)
Half the US reactors	1200	7000
Half of all reactors in US and Europe	2400	14,300
All reactors in US, Europe, and Japan	4800	28,600
All reactors in Russia	400	2600
All reactors in Russia, FSU, and China	1600	9500
All reactors in US, Europe, Japan, Russia, FSU, and China	6300	38,000

2. Background

Thorium was examined as a fuel type in many countries in the 1950s, 1960s, and 1970s. The United States irradiated thorium fuel in the 1950s in the Shippingport light water breeding reactor. The reactor was fueled with ^{233}U and ^{232}Th and produced 1.4% more fuel than it burned (Olson et al., 2002). Other reactors such as Indian Point I and Ft. St. Vrain used thorium fuel as well.

Thorium fuel has different neutronic properties than uranium fuel. Natural thorium (Th) consists solely of ^{232}Th, which is a fertile but not fissile isotope. Fertile means that it does not fission, but the isotope can absorb neutrons and transmute into a fissile isotope. Thorium undergoes neutron absorption and two successive beta decays to produce fissile ^{233}U. Traditional uranium oxide fuel is enriched in the isotope ^{235}U, which is also fissile. The major isotopic component of uranium, ^{238}U, may fission at high energies, but it is also fertile and produces the fissile plutonium isotopes ^{239}Pu and ^{241}Pu. The number of neutrons produced per neutron absorbed (η) is higher for ^{233}U than for other fissile isotopes in a thermal spectrum (see Table 2), which makes it attractive for power production with minimal actinide buildup. The absorption cross section of ^{232}Th is larger than that of ^{238}U, which means that thorium generates the fissile isotope ^{233}U more efficiently than uranium produces fissile plutonium. Additionally, the final product of irradiated thorium fuel sent to a repository consists of actinides that pose a reduced radiotoxicity for the first 10,000 years, but thorium by-products such as radium lead to a slight increase in radiotoxicity compared to UO_2 fuel from 10,000 to 1 million years after removal from the reactor (Galperin et al., 2002 and Trellue et al., 2010).

Because the fissile isotope ^{233}U must be produced during irradiation, a nuclear power reactor cannot sustain criticality with just thorium fuel; it must at least initially contain other fissile material (e.g., low-enriched uranium (LEU) and/or plutonium). As ^{233}U builds into a Th/LEU system, the average delayed neutron fraction and the recoverable energy per fission decrease (see Table 2). The delayed neutron fraction (β) is an important safety parameter because it indicates how much of an impact changes to the system

Table 2
Neutronic properties of fissile isotopes examined (Lamarsh, 1983; Knief, 1992, and Wilson et al., 1995).

Isotope	η	ν	β	Q
U-233	2.287	2.492	0.0026	200.29
U-235	2.068	2.418	0.0065	202.61
Pu-239	2.108	2.871	0.0021	211.41
Pu-241	2.145	2.927	0.005	213.41

Table 3
Summary of fuel compositions considered.

Case	Fuel composition	Calc. type	Fuel
1	94% Th, 6% WGPu	Full core	Th-WGPu
2	89% Th, 5% NU, 6% WGPu	Full core and Pin-cell	Th-WGPu, denatured
3	88% Th, 6% NU, 6% WGPu	Pin-cell	Th-WGPu with increased denaturing
4	90% Th, 10% RG-Pu	Pin-cell	Th-RG-Pu, no denaturing
5	85% Th, 5% NU, 10% RG-Pu	Pin-cell	Th-RG-Pu, Denatured
6	84% Th, 6% NU, 10% RG-Pu	Pin-cell	Th-RG-Pu with increased denaturing
7	80% Th, 20% U(19.9% ^{235}U)	Full core	Th-LEU
8	100% U(4.2% ^{235}U)	Full core	LEU
9	94% Th, 6% WGPu, higher burnup	Pin-cell	Th-WGPu with burnup = 94 MWd/kg
10	92% Th, 2% NU, 6% WGPu, higher burnup	Pin-cell	Th-WGPu, slight denaturing and higher burnup
11	89% Th, 5% NU, 6% WGPu, higher burnup	Pin-cell	Th-WGPu, denatured with higher burnup
12	80% Th, 20% U(19.9% ^{235}U)	Pin-cell	Th-LEU with higher burnup
13	94% DU, 6% WGPu	Full core	MOX with WGPu
14	90% DU, 10% RG-Pu	Pin-cell	MOX with RG-Pu

make as a function of time (Note that similar neutronic-based safety issues occur when ^{239}Pu-based fuels are introduced in a reactor because the delayed neutron fraction of ^{239}Pu is similar to that of ^{233}U). Control rods and soluble boron are similarly less effective in decreasing reactivity in ^{233}U- and ^{239}Pu-fueled systems than in ones containing only ^{235}U. The smaller recoverable energy per fission of ^{233}U means that slightly more fuel must be burned to maintain the same power level as enriched uranium fuel. Thorium may exist within its own assemblies in portions of the core, or it may be heterogeneously dispersed within fuel rods and/or assemblies containing UO_2 fuel (such as the Radkowsky Thorium Fuel concept) or PuO_2 (Galperin et al., 2002). Combining thorium with either weapons- or reactor-grade plutonium provides an excellent Pu destruction mechanism for the latter.

Preliminary neutronic and thermal hydraulic calculations in both PWRs and BWRs showed that reactors can safely burn certain amounts of thorium fuel (Dziadosz, et al., 2004, Kazimi et al., 1999, and Shwageraus et al., 2004b). A significant presence of either ^{233}U or ^{239}Pu decreased some of the safety margins compared to LEU, which meant that full cores of either MOX or thorium/plutonium fuel would be more difficult to implement than traditional uranium fuel. Reactor design modifications were required to burn a full core

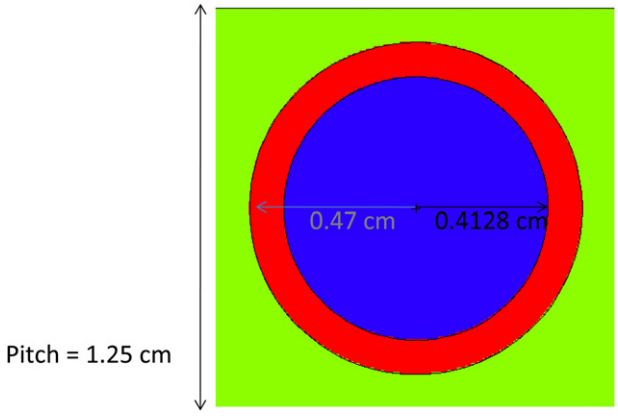

Fig. 1. Geometry used for PWR pin-cell.

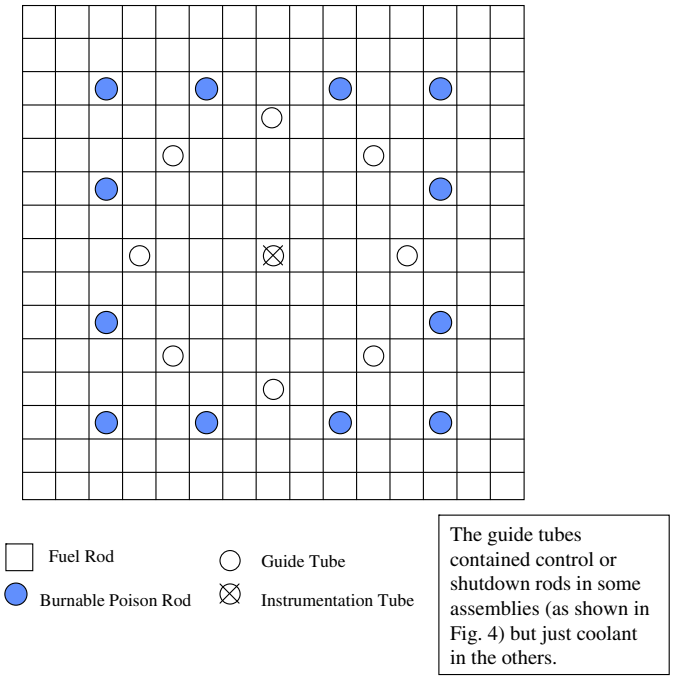

Fig. 2. Fuel pin layout in an H. B. Robinson 15 × 15 PWR assembly.

of MOX fuel (Trellue, 2004); similar ones would be applied to thorium/plutonium or high burnup thorium/uranium fuel when a majority of the fissions come from ^{233}U or ^{239}Pu. Other studies showed that thorium fuel was beneficial for plutonium destruction and that denaturing the fuel with uranium made the resulting uranium feed stream less attractive (Bathke et al., 2008). Designs for systems with separate uranium and thorium fuel sections (i.e., seed and blanket) have been developed to increase burnup in the thorium, but hot spots and thermal parameters may prevent implementation of such designs. Systems with duplex pellets (i.e. 19.9% enriched UO_2 in the center portion of the fuel surrounded by thorium) have also been examined but posed challenges with thermal properties (Shwageraus et al., 2004a). Other studies used Monte Carlo techniques for neutronic studies of a single thorium fuel pin, such as the radial profile of ^{233}U buildup (Herring et al., 2004), but not for a larger core model.

Whereas neutronic and reactivity safety calculations have been performed for thorium fuel in the past, material attractiveness represents a new component of fuel cycle analyses. Material attractiveness calculations are performed with Monte Carlo techniques; thus, using Monte Carlo to perform burnup calculations simplifies the interface between the two.

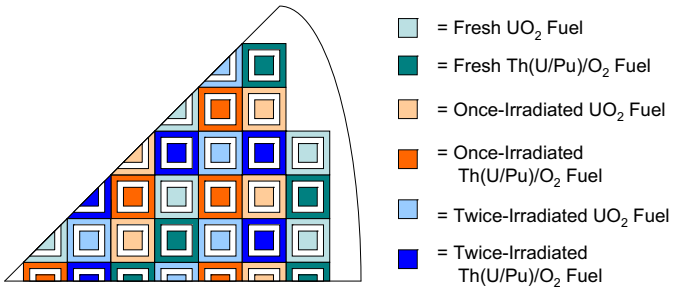

Fig. 3. Layout of assemblies within the PWR core for a partial Th loading (note the squares represent three possible burnup regions for each assembly).

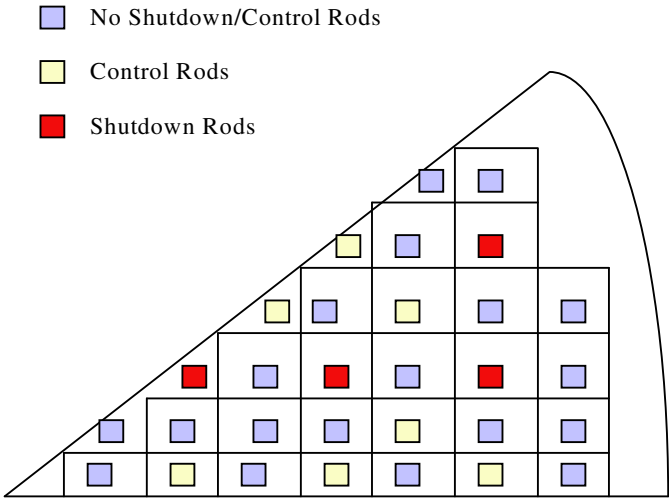

Fig. 4. Location of assemblies with control/shutdown rods for safety calculations performed for PWR core.

3. Method

Three different types of calculations were performed as part of this research and are discussed in the following sections: 1) burnup (changes in isotopics as a function of irradiation), 2) reactivity safety, and 3) material attractiveness.

3.1. Burnup calculations

The purpose of burnup calculations was to compare the reactivity and material compositions as a function of irradiation between various fuel types. The fuel types considered were: low-enriched uranium (LEU), traditional plutonium/uranium mixed oxide (MOX) fuel, thorium mixed with low-enriched uranium (19.9%), thorium mixed with plutonium, and thorium mixed with both plutonium and natural uranium (to make the uranium isotopic vector less attractive upon discharge). To develop a fundamental understanding of the reactivity changes for a large number of different starting material compositions efficiently, a representative pin was irradiated. The enrichments of fissile material in the fuel were randomly chosen and were not expected to have comparable reactivity-limited discharge burnup. Simply a fixed burnup of 47 GWd/MTU was chosen to represent fuel irradiated in a PWR for three cycles; additionally, a few cases were run with a theoretically doubled burnup (i.e., 94 GWd/MTU). Specific calculations using a model with a reflected 1/8 core model where each assembly was modeled as a separate burnup region and shuffling of assemblies occurred were then run for select cases. The code *Monteburns* linked the Monte Carlo transport code MCNP with the isotope generation and depletion code ORIGEN2.2 and was used in this research to calculate reactivity and isotopic concentrations as a function of burnup (Trellue and Poston, 1999). One benefit of using Monte Carlo codes such as MCNP and *Monteburns* compared to deterministic codes was that they allowed flexible geometry modeling with system-dependent, infinite dilute, temperature-dependent cross sections, eliminating the need for problem-dependent, multi-group shielded cross sections. The disadvantage of using Monte Carlo statistical solutions was that a large amount of computer time and memory was required to complete a complex calculation. Previously, calculations using Monte Carlo techniques were limited to less than about forty burn materials (i.e., typically a fuel pin or assembly with specified fuel regions). To use *Monteburns* to simulate burnup of an entire PWR core, pins were modeled individually but were grouped for burnup calculations. With advances in computing power, the number of regions that can be modeled constantly increases. For the results presented in this paper, only a few cases were performed using the 1/8 core model to show the capability existed; the majority were examined using a simple pin-cell model of a PWR fuel rod and the surrounding water because the pin-cell offered a good comparison between fuel types.

Table 3 summarizes the cases considered in this study, both pin-cell and full core models. The plutonium cases were chosen because surplus weapons-grade plutonium currently exists in the United States and an efficient disposition method is needed. Similarly, reactor-grade plutonium exists within used fuel assemblies, and without a permanent repository for used fuel, transmutation of the material may become desirable. The LEU/thorium case was chosen because it provided an unattractive uranium discharge stream and was the basis of initial thorium fuel cycle studies. The best plutonium destruction rate was obtained when just thorium and plutonium fuel was used; the purpose of adding a little natural uranium in some cases was to decrease the material attractiveness of the uranium product after irradiation. Although thorium cases were the focus of this research, both low-enriched uranium (LEU) and uranium–plutonium mixed oxide (MOX) fuel were studied for comparative purposes. The amount of plutonium in the fuel was estimated based on previous research (Trellue, 2004).

Table 4
Details of fuel assembly modeled.

Parameter	Material	Density	Temperature	Radius
Reactor-grade plutonium	4.078 wt% ^{238}Pu, 47.61 wt% ^{239}Pu, 26.20 wt% ^{240}Pu, 13.48 wt% ^{241}Pu, 8.625 wt% ^{242}Pu;	10.05 g/cm^3	900 K	0.4647 cm
Weapons-grade plutonium	93 wt% ^{239}Pu, 7 wt% ^{240}Pu,	10.05 g/cm^3	900 K	0.4647 cm
Cladding	Zircaloy-4	6.44 g/cm^3	600 K	0.4742–0.5360 cm
Coolant	Light Water	0.737 g/cm^3	600 K	Pitch – 1.43 cm
Burnable poison	Borosilicate glass	2.23 g/cm^3	600 K	0.3086–0.5029 cm

Table 5
Time steps for burnup calculations.

Time		Soluble boron concentration
0.5 days	Irradiation	652.5 ppm
1.0 days	Irradiation	652.5 ppm
5.5 days	Irradiation	652.5 ppm
90.14 days	Irradiation	652.5 ppm
90.14 days	Irradiation	652.5 ppm
90.14 days	Irradiation	652.5 ppm
90.14 days	Irradiation	326.25 ppm
90.14 days	Irradiation	326.25 ppm
90.14 days	Irradiation	326.25 ppm
64 days	Decay	

Table 6
The meaning of FOM when applied to metals or alloys.

FOM	Weapons utility	Attractiveness	Attractiveness level (Nuclear Material Control and Accountability, 2006)
>2	Preferred	High	~B
1–2	Attractive	Medium	~C
0–1	Impractical	Low	~D
<0	Very impractical	Very low	~E

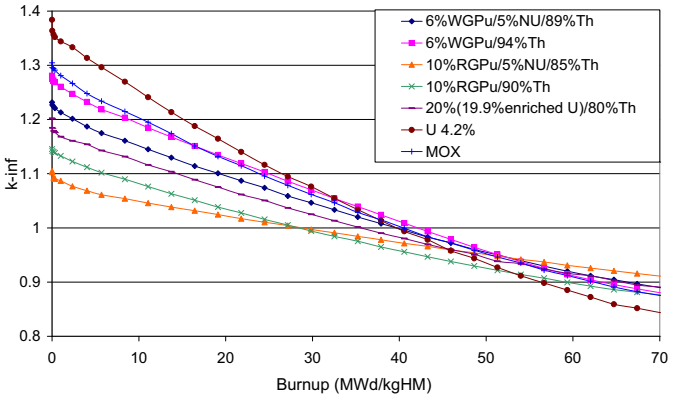

Fig. 5. Reactivity results from pin-cell calculations.

3.1.1. Pin-cell calculations

Initial neutronic calculations were performed using an infinitely-reflected pin-cell calculation with fuel, cladding, and water representing a PWR (shown in Fig. 1). A pin-cell calculation provided a good estimation of reactivity and isotopic changes as a function of burnup. However, it was not representative of the exact conditions a fuel rod undergoes in a reactor because its reactivity was seldom critical. Within a reactor, the core itself is kept at critical, but the reactivity in a particular pin may be either supercritical or subcritical. Control measures ensure that criticality is maintained in the system as a whole as a function of burnup. Thus, in the pin-cell calculations, k_{eff} neither started nor ended at exactly 1.0.

3.1.2. Full core calculations

To examine reactivity safety coefficients and other aspects of thorium-based fuel, more detailed calculations were required during which the spectrum of the system remained closer to critical throughout burnup. Thus, a one-eighth core of an H. B. Robinson PWR (Hermann et al., 1995) using MCNP and *Monteburns* was used to simulate a PWR undergoing multiple irradiation cycles. Each assembly contained fuel rods, burnable poison rods, and guide tubes into which control and/or shutdown rods were inserted when applicable (see Fig. 2). All fuel rods within each assembly were either modeled as one big fuel region or were separated into three assembly-level regions for burnup. The three regions included: the outer "box", the inner square, and the middle (see the "white" boxes in Fig. 3 where half the fuel was UO_2 and half was Th/PuO_2). For safety calculations, control or safety rods were placed in some of the assemblies; others simply contained guide tubes (see Fig. 4). All cases assumed that one-third of the core was reloaded and the other two-thirds was shuffled every year and half (shuffling was modeled using a feed input file in *Monteburns*). Each fuel rod was modeled as the same material in the axial dimension (365.76 cm). To model each assembly in a 1/8 core representation of a PWR, at least thirty different materials were required (ten assemblies for each of three different irradiation loadings) although many were only partial assemblies around the infinitely-reflective boundaries. Ninety fuel materials were burned in cases with three regions per assembly.

The power of this 1/8 core was 253 MWt to simulate a 2000 MWt H. B. Robinson reactor. The innermost (center) 1/8 of an assembly was assumed to be thrice-irradiated fuel. The most accurate technique of following fuel in the reactor for three cycles was to already have irradiated fuel placed in the two-thirds of the system not currently being loaded. Initial isotopic estimates of the two-thirds of the reactor that were not fresh fuel (i.e., once- and twice- irradiated fuel) were obtained using results of pin-cell burnup calculations. By cycle 3, the fuel materials in all regions started representing an equilibrium state. Thus, a total of five cycles of irradiation were required to follow fresh fuel loaded at the beginning of cycle 3 through this "equilibrium" system. In most cases, each cycle consisted of ten burn steps with small steps at the beginning to capture xenon buildup in the first couple of days of irradiation. Table 4 contains details of the assembly modeled for these calculations, and Table 5 gives the time steps used for irradiation. The soluble boron concentration in the water started at 652.5 ppm and was reduced by a factor of two about halfway through each irradiation cycle. The densities of both Pu/Th oxide and UO_2 were both 10.0 g/cc for this study although ThO_2 would probably have a lower density when actually fabricated. The reactivity ranged from a k_{eff} of about 1.07 at the beginning of a cycle to 1.0 at the end of a cycle; in reality, daily soluble boron adjustments would make the reactor critical throughout each cycle.

3.2. Reactivity safety calculations

Kinetic safety parameters were calculated using the 1/8 core models created as described in Section 3.1.1. The boron efficiency was calculated by changing the boron concentration in the water coolant by 50–100 parts per million (ppm) less and 50–100 ppm more than the default case in two different MCNP runs. The initial boron concentration (in ppm) was the amount required to keep the reactor at critical at the beginning of an equilibrium cycle and was calculated but not optimized for each case. Both control and shutdown rods were located in clusters, which in this case were comprised of nine rods each and occupied the guide tube locations of certain assemblies. The assemblies containing control or shutdown rod clusters (located in the nine guide tubes of the assembly) are given in Fig. 4. The control and shutdown rod worths were obtained by calculating the difference between k_{eff} when control and/or shutdown rod materials were added to the appropriate guide tube locations and the default case. The total rod worth was that which occurred when both control and shutdown rods were inserted.

Appendix A provides results of an assembly-level benchmark calculation for a PWR performed with *Monteburns* to a calculation performed with the code package CASMO/SIMULATE (Bjork and

Table 7
Details of full core calculations.

Case		1st half	2nd half	Type of model
1	Half core of Th/Pu	6% WGPu, 94% Th	4.2 wt% UO_2	90-region
2a	Half core of Th/U/Pu	6% WGPu, 5% NU, 89% Th	4.2 wt% UO_2	30-region
2b	Full core of Th/U/Pu	6% WGPu, 5% NU, 89% Th	6% WGPu, 5% NU, 89% Th	30-region
7	Half core of Th/LEU	19.9 wt% UO_2, 80% Th	4.2 wt% UO_2	90-region
8	Full core of LEU	4.2 wt% UO_2	4.2 wt% UO_2	90-region
13a	Half core of MOX	6% WGPu, 94% NU	4.2 wt% UO_2	30-region
13b	Full core of MOX	6% WGPu, 94% NU	6% WGPu, 94% NU	30-region

Table 8
Change in fissile material inventory: tons/18-month discharge in 50 3400 MWt reactors.

#	Initial fuel composition(~1500 tons of actinide)	MWd/kg	Initial fissile	^{233}U	^{239}Pu	^{241}Pu	Total	^{235}U
1	94% Th, 6% WGPu	47	83.7	20.1	−75.3	6.9	−48.3	
2	89% Th, 5% NU, 6% WGPu	47	83.7	20.1	−69.0	7.6	−41.3	
3	88% Th, 6% NU, 6% WGPu	47	83.7	21.7	−68.9	7.3	−39.9	
4	90% Th, 10% RG-Pu	47	91.6	22.9	−55.7	−5.8	−38.6	
5	85% Th, 5% NU, 10% RG-Pu	47	91.6	21.7	−49.4	−1.7	−29.4	
6	84% Th, 6% NU, 10% RG-Pu	47	91.6	22.4	−49.3	−5.2	−32.1	
7	80% Th, 20% U (19.9% ^{235}U)	47	60	18.2	2.4	0.8	21.4	−48.6
8	100% U(4.2% ^{235}U)	47	63	0.0	8.4	2.4	10.8	−52.2
9	94% Th, 6% WGPu	94	83.7	24.7	−83.2	1.1	−57.4	
10	92% Th, 2% NU, 6% WGPu	94	83.7	25.8	−82.3	2.4	−54.1	
11	89% Th, 5% NU, 6% WGPu	94	83.7	26.0	−81.6	2.9	−52.7	
12	80% Th, 20% U (19.9% ^{235}U)	94	60	21.8	3.5	1.1	26.4	−57.2
13	MOX WGPu	47	83.7	0.0	−51.1	8.8	−42.3	
	1/3 core MOX WGPu/2/3 core UO2	47	70	0.0	−11.4	4.5	−6.9	−34.8
14	MOX RG-Pu	47	91.6	0.0	−28.9	−3.8	−32.7	
	1/3 core MOX RG-Pu/2/3 core UO2	47	72.5	0.0	−4.0	0.3	−3.7	−34.8

Fhager, 2009). The study showed that similar results were obtained using each technique.

3.3. Attractiveness calculations

A comparison of the material attractiveness associated with each of the fourteen cases in Table 3 was made using the Figure of Merit (FOM) formulation (Bathke et al., 2009). The FOM was formulated using the primary factors of material attractiveness (*i.e.*, the bare critical mass, the internal heat generation, and the radiation dose rate (Beller and Krakowski, 1999)), so that the formula cannot be reverse engineered to reveal weapons design information. The FOM_1 given in Equation [1] was used herein, because it indicated the worst-case threat. FOM_1 applied to a technically advanced proliferant nation (in purified metal form) or a dedicated sub-national group (in unpurified metal form). Material attractiveness was calculated for spent-fuel compositions derived from both pin-cell and full core results.

$$FOM_1 = 1 - \log_{10}\left[\frac{M}{800} + \frac{Mh}{4500} + \frac{M}{50}\left[\frac{D}{500}\right]^{\frac{1}{\log_{10}2}}\right] \quad (1)$$

- M – bare critical mass in purified or unpurified metal form (kg)
- h – heat content in purified or unpurified metal form (W/kg)
- D – dose rate of 0.2 M @ 1 m (rad/h)

Table 6 gives the meaning of FOM values in terms of weapons utility, and materials attractiveness in a safeguards and security context. To make a material unattractive for use in a nuclear device, its FOM value must be less than 1. Table 6 reflects the fact that while a particular nuclear material might be preferable for use in a nuclear weapon or explosive device, the design and construction of effective nuclear weapons from any of the materials with $FOM_1 > 1$ is potentially usable and for materials with $FOM_1 < 1$ is impractical, but still may be theoretically possible. The fact that potential proliferant states or sub-national groups might "prefer" one material over another should not imply that either material in question is "proliferation-proof," or that any reduction in international safeguards and national physical protection requirements can be justified. It should be noted, however, that the lower the FOM_1 the better. Even though a material may still need to be safeguarded and secured, a process that produces a material with a FOM value of 1.1 should be encouraged over a process that produces a material with FOM value of 2.5.

4. Results

Although much information can be inferred about thorium fuel, this research focuses on neutronic burnup, safety, and material attractiveness calculations. Results from neutronic calculations are given in Section 4.1. Neutronic safety (kinetic) results are presented in Section 4.2, and material attractiveness is discussed in Section 4.3.

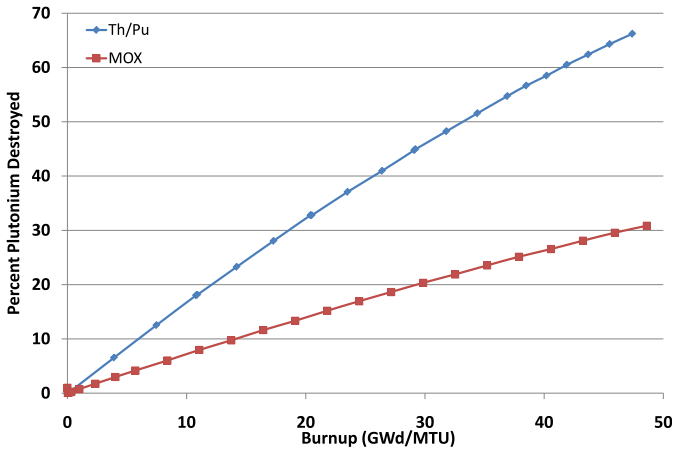

Fig. 6. Percent of Pu destroyed using weapons-grade Th/Pu and U/Pu (MOX) fuels.

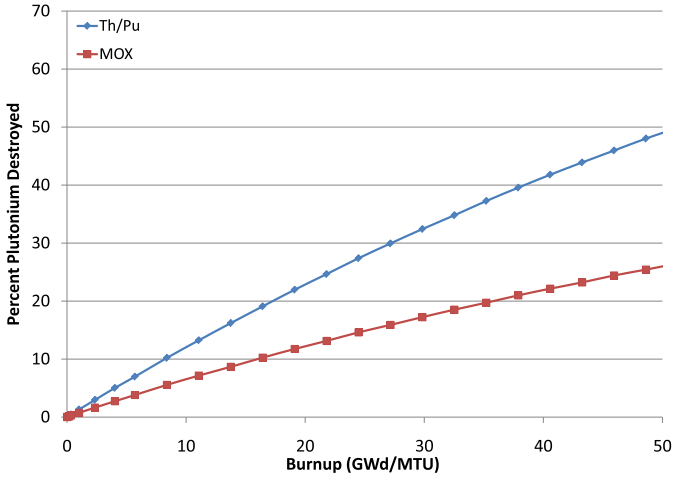

Fig. 7. Percent of Pu destroyed using reactor-grade Th/Pu and U/Pu (MOX) fuels.

Table 9
Kinetic safety parameters for U/Th case compared to UO_2.

	UO_2		LEU/Th in half the core	
	BOL	EOL	BOL	EOL
β_{eff}	0.00628	0.00519	0.00595	0.00504
Boron efficiency (pcm/ppm)	9.1	9.6	8.8	8.9
Total rod worth (pcm)	5735	5672	5014	5517
Control rod worth (pcm)	3838	3073	2900	3073
Initial boron concentration (ppm)	1800		600	

4.1. Neutronic results

Neutronic calculations of various types of system models were performed as described in Section 3, including a pin-cell, a full core with 90 fuel materials (three regions per assembly as shown in Fig. 3), and a full core with 30 fuel materials (one material per assembly).

All cases listed in Table 3 were run as pin-cell calculations; some were additionally run as full core cases as well. Reactivity changes for selected results are given in Fig. 5. Results from pin-cell cases were taken at both 47 and 94 MWt d/kgHM, the former being taken to correspond more directly with the full core cases, and the latter being a theoretically-achievable burnup that would require significant material and design changes to achieve. Cladding failures and other mechanical issues typically exist for burnups of over 60 MWt d/kgHM with oxide fuels and zircaloy cladding. New cladding such as silicon carbide (SiC) fiber-reinforced SiC composites, or oxide dispersion-strengthened steels would need to be examined to allow higher burnups.

Thorium cases were generally associated with smaller initial reactivity than low-enriched uranium ones, but the reactivity of thorium-based fuels remained flatter at higher burnups (i.e. greater than 50 MWt d/kgHM). The inclusion of natural uranium in the thorium/plutonium fuel decreased the reactivity at the beginning of life (BOL) because it was a neutron absorber. However, the presence of natural uranium increased reactivity at higher burnups because it bred in ^{239}Pu. Reactor-grade plutonium did not provide as large of an initial reactivity as weapons-grade plutonium because it contained less fissile plutonium, but it produced the most self-sustaining neutron economy during irradiation, particularly when mixed with natural uranium (NU) initially.

To provide more realistic PWR reactivity and kinetic safety calculations, a model more representative of a full core was necessary. Some of the fuel mixtures studied as pin-cell calculations were incorporated into full core models of the core as either full or half cores of thorium-based fuel (see Table 7). After three cycles of irradiation, neutronic results for the spent nuclear fuel from the one-eighth core cases represented an average burnup of about 47 MWt d/kgHM. Further calculations using the 1/8 core model indicated that to burn thorium-based fuel to 94 MWt d/kgHM, each fuel assembly would have to stay in the reactor for approximately six cycles instead of three. Additionally, approximately one-half of the core would have to be 5.5–6 wt% enriched uranium irradiated for the usual three cycles to maintain sufficient reactivity to counteract the effects of high burnup thorium fuel in other locations of the core. Table 8 gives the change in fissile isotope inventories for each case. In a typical PWR operating at 3400 MWt, the amount of fuel in the system at any time was about 90 tons. Table 8 results were obtained by assuming that 1/3 of the core (i.e. 30 tons) was discharged at each reloading (i.e., every 18 months) in each of 50 different reactors, which was approximately 1/2 of all reactors in the United States. The amount of plutonium destroyed using MOX versus Th/Pu fuel as a function of irradiation is shown in Figs. 6 and 7.

4.2. Reactor kinetic safety parameters

To determine how a reactor responds to changes in the temperature and materials present in the system, neutronic safety parameters were calculated (see Tables 9–11). Parameters such as the Doppler fuel and moderator temperature coefficients exhibited expected behavior and are not shown. The boron efficiency represented how much of an impact changes in the soluble boron concentration had on reactivity. References (Youinou et al., 1999) suggest that in a PWR, boron efficiency should be greater than 4 pcm/ppm (pcm is a change in k_{eff} of 1.0e-5, and ppm represents parts per million of boron). The total rod worth indicated how quickly the reactor responded when control and shutdown rods were inserted and should be greater than 5000 pcm (Youinou et al., 1999). By allowing only half the core to be thorium-based fuel and the other half to be traditional low-enriched uranium oxide, the boron efficiency remained greater than 4 pcm/ppm and the control rod worth greater than 5000 pcm. However, the magnitude of both of these values was not as large for thorium-based fuel as a full core of UO_2 because differences in the flux spectrum and absorption capability of the system made boron less efficient. Results for boron efficiency and total rod worth for full cores of thorium fuel were below the recommended values as were values for full cores of MOX fuel in a PWR. However, the real constraints are not just a fixed number; they depend on how much reactivity needs to be suppressed, which is influenced by water chemistry and other coolant conditions. One concern was that the initial boron concentration is too large in both the thorium/plutonium and MOX cases and may require slight design changes.

These studies indicated that thorium posed some of the same concerns as the use of traditional plutonium-uranium mixed oxide (MOX) fuel in a reactor. The delayed neutron fraction of ^{239}Pu was similar to that of ^{233}U, and the resulting boron efficiencies and control and shutdown rod worths decreased as either isotope built in. A previous study examined the use of full cores of MOX fuel in a PWR and made the following suggestions to make the cores "safer": 1) increased soluble boron enrichment to at least 25 wt% ^{10}B (natural boron is 19.9% enriched in ^{10}B), 2) changed the composition of both control and shutdown rods to enriched B_4C (standard control rods are a Ag–In–Cd alloy), and/or 3) added extra

Table 10
Kinetic safety parameters for half cores of plutonium-based fuel.

	WGPu/Th in half the core		WGPu/U/Th in half the core		WGPu/MOX in half the core	
	BOL	EOL	BOL	EOL	BOL	EOL
β_{eff}	0.00486	0.00404	0.00463	0.00392	0.00502	0.00439
Boron efficiency (pcm/ppm)	6.8	7.2	6.5	6.8	6.8	6.9
Total rod worth (pcm)	5318	5404	5521	4473	5710	5437
Control rod worth (pcm)	2941	3017	3153	3245	3273	3038
Initial boron concentration (ppm)	1100		2000		2666	

Table 11
Kinetic safety parameters for full cores of plutonium-based fuel.

	WGPu/Th in a full core		WGPu/U/Th in a full core		WGPu/MOX in a full core	
	BOL	EOL	BOL	EOL	BOL	EOL
β_{eff}	0.00318	0.00313	0.00323	0.00340	0.00377	0.00407
Boron efficiency (pcm/ppm)	4.7	5.3	4.4	4.9	4.4	4.8
Total rod worth (pcm)	4816	5061	4923	4852	5071	5049
Control rod worth (pcm)	2562	2642	2783	2602	2757	2759
Initial boron concentration (ppm)	2900		2666		2800	

Table 12
Material attractiveness for each case.

Case	Fuel mix	Plot label	Burnup (MWd/kg)	FOM_1	
				Pu	U
1	94% Th, 6% WGPu	wgpu	47	2.16	2.46
2	89% Th, 5% NU, 6% WGPu	upu	47	2.29	1.54
3	88% Th, 6% NU, 6% WGPu	wgupu5	47	2.10	1.48
4	90% Th, 10% RG-Pu	rgpu	47	1.59	2.47
5	85% Th, 5% NU, 10% RG-Pu	rgupu	47	1.77	1.60
6	84% Th, 6% NU, 10% RG-Pu	rgpu3	47	1.66	1.50
7	80% Th, 20% U (19.9% 235U)	thu	47	1.84	0.87
8	100% U (4.2% 235U)	u42	47	2.09	–
9	94% Th, 6% WGPu	wgpu2	94	1.16	2.29
10	92% Th, 2% NU, 6% WGPu	wgupu3	94	1.33	2.01
11	89% Th, 5% NU, 6% WGPu	wgupu2	94	1.41	1.68
12	80% Th, 20% U (19.9% 235U)	thu2	94	1.56	0.89
13	MOX WGPu	moxw	47	2.26	–
14	MOX RG-Pu	moxr	47	1.81	–

water holes or decreased fuel rod diameter to increase the moderator-to-heavy metal ratio (Trellue, 2004).

4.3. Attractiveness results

Table 7 gives the calculated material attractiveness of the irradiated plutonium and uranium product for each of the 14 cases examined, assuming the used fuel was reprocessed five years after discharge to provide separated plutonium and uranium end products. The least attractive material has a value of $FOM_1 << 0.0$, which is why there was no result for FOM_1 for uranium in irradiated uranium oxide or mixed oxide fuel. The addition of small quantities of natural uranium (NU) in the thorium/plutonium fuel charge (denaturing) made the final uranium product less attractive than non-denatured material, as seen by comparing Case 1 with 2 and 3, Case 4 with 5 and 6, and Case 9 with 10 and 11. However, the effect that the additions of NU had on the final plutonium product varied depending on the burnup and grade of the initial plutonium: for Cases 9, 10, and 11, the final plutonium product was more attractive with increasing initial NU concentration; and for Cases 1 through 6, the attractiveness of the final plutonium product increased and then decreased with increasing initial NU concentration.

Increasing the burnup from 47 to 94 MWt d/kgHM significantly decreased the attractiveness of weapons-grade plutonium (WGPu), as seen by comparing Case 1 with 9 and Case 2 with 11. The effect of increasing the burnup on the attractiveness of the final uranium product depended on the initial concentration of NU. Increasing the burnup from 47 to 94 MWt d/kgHM also decreased the attractiveness of the reactor-grade plutonium (RG-Pu) product, as seen by comparing Case 7 with 12, but the attractiveness of the final uranium product increased slightly.

As can be seen from Table 12, FOM_1 was greater than 1 for the plutonium in the used fuel for all 14 cases and for the uranium in the used fuel for 2/3 of the 14 cases. One way to render the plutonium or the uranium unattractive to a non-Host State adversary was to co-extract these elements with the thorium in the used

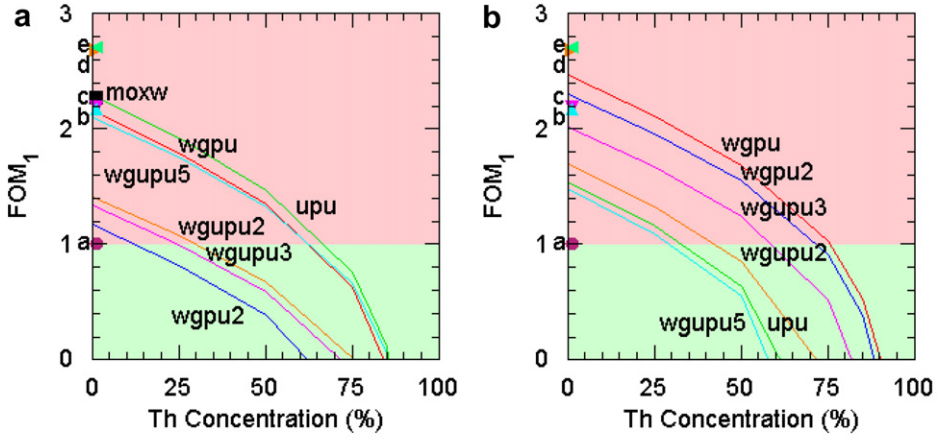

Fig. 8. a/b. The attractiveness of a Th + Pu mixture (left) and Th + U mixture (right), respectively, from 5-year old Th used fuel with WGPu seed.

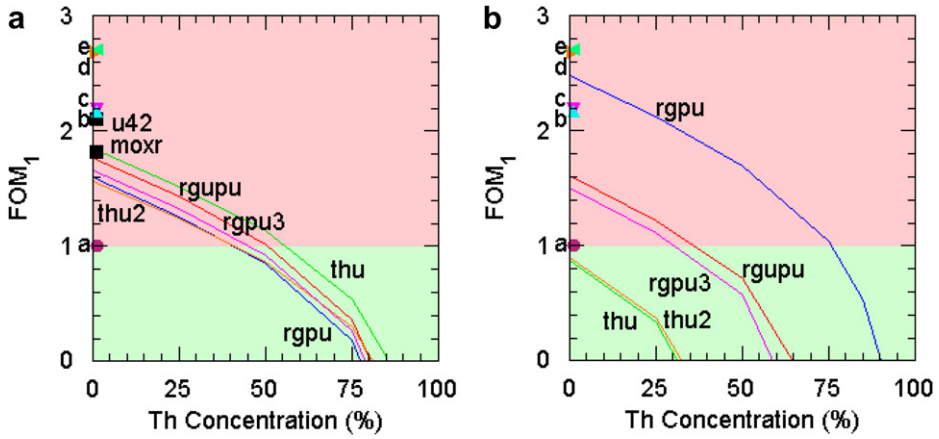

Fig. 9. a/b. The attractiveness of a Th + Pu mixture (left) and Th + U mixture (right), respectively, from 5-year old used fuel with RG-Pu seed.

Table 13
Kinetic safety parameters for thorium/plutonium fuel using *Monteburns* versus casmo/simulate (add reactivity as a function of burnup).

BOL	RG-Pu	AmPu	WGPu	2RPu
Boron efficiency (MB)	3.14	3.42	4.12	2.11
Boron efficiency (CASMO)	2.9	2.5	3.6	2.5
Moderator coefficient (MB)	−42.96	−37.47	−50.44	−32.27
Moderator coefficient (CASMO)	−46	−46	−45	−38
Doppler coefficient (MB)	−4.72	−3.92	−4.84	−3.91
Doppler coefficient (CASMO)	−3.4	−3.25	−3.45	−3.4
EOL				
Boron efficiency (MB)	3.94	2.49	4.77	2.27
Boron efficiency (CASMO)	3.8	3.0	5.7	3.0
Moderator coefficient (MB)	−37.13	−33.36	−36.95	−31.53
Moderator coefficient (CASMO)	−48	−47	−37	−47
Doppler coefficient (MB)	−3.50	−2.74	−3.48	−3.59
Doppler coefficient (CASMO)	−3.5	−3.4	−3.45	−3.5

fuel. The thorium concentration that was required to render a Th + Pu or a Th + U mixture unattractive for the cases that had initially been seeded with WGPu is shown in Fig. 8a and b respectively. Similarly, the thorium concentration that was required to render a co-extracted Th + Pu or a Th + U mixture unattractive to a non-Host State adversary for the cases that had initially been seeded with RG-Pu is shown in Fig. 9a and b respectively.

5. Conclusions and future work

Thorium is becoming a part of the worldwide nuclear fuel cycle because it is more abundant in some countries than other energy resources and is thus important to characterize. Results from this study confirmed that thorium/plutonium fuel can consume plutonium more efficiently than traditional uranium/plutonium MOX fuel. Some design changes may be required for full cores of thorium/plutonium fuel to operate safely compared to traditional UO_2, but quantities of plutonium present today could indeed be effectively burned in existing Light Water Reactors. Additionally, if technically feasible, increasing the burnup of thorium/plutonium fuel to 94 MWt d/kgHM both increased the plutonium consumption further and decreased the material attractiveness of the plutonium. Denaturing the thorium/plutonium fuel by adding natural uranium at charge decreased the material attractiveness of the uranium. However, the "optimal" amount of uranium that should be added still needs to be addressed.

This paper showed that Monte Carlo analysis can produce similar results to deterministic codes for the use of thorium fuels in a PWR and as computational resources become more powerful, detailed full core burnup analyses of PWRs may become possible. The results of full core neutronics calculations in this paper in which all rods in an assembly were grouped as one material region represented an approximation of actual fuel irradiation shuffling in which every fuel pin in the reactor undergoes a different flux, but they represent an upcoming era of more complex Monte Carlo computations. Incorporating burnup results to material attractiveness calculations yields additional information about new fuel types not previously available.

Appendix A. Benchmark comparison of *Monteburns* to casmo/simulate for thorium/plutonium fuel in a pressurized water reactor

Three different kinetic safety parameters were analyzed: boron worth, Doppler fuel temperature coefficient, and the moderator temperature coefficient both at the beginning and end of life (~46.6 GWd/MTHM). Results are given in Table 13.

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